

REMARKS

This document is submitted in reply to the Office Action dated February 13, 2009 (Office Action).

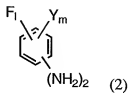
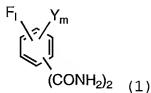
Applicants have amended claims 1, 5, 6, 11, and 12 to more particularly point out their invention. Support for these amendments can be found in the Specification at page 9, line 2 and in previously presented claim 13. Applicants have also canceled claim 13. Finally, Applicants have added a new claims, i.e., claims 16-20, to cover a particular embodiments. No new matter has been introduced.

Upon entry of these amendments, claims 1-12 and 14-20 will be pending and under examination. Applicants respectfully request that the Examiner reconsider this application, as amended, in view of the following remarks.

Rejection under 35 U.S.C. § 103

Claims 1-15 are rejected as obvious over Masayoshi et al., European Patent Application No. 1275679 ("Masayoshi"), in view of Andrews et al., Aust. J. Chem., 1971, 412-422 ("Andrews"), Hazen et al., U.S. Patent No. 5,011,997 ("Hazen"), and Pfirrmann et al., U.S. Patent No. 3,897,498 ("Pfirrmann") as evidenced by Zengel et al., U.S. Patent No. 5,410,082 ("Zengel"). See the Office Action, page 2, lines 16-23.

Independent claim 1 will be discussed first. This claim, as amended, covers a method for obtaining a fluorinated phenylenediamine of formula (2) shown below. The diamine of formula (2) is obtained via the Hoffman rearrangement by reacting a diamide, represented by formula (1) below, with NaOX, where X is either Br or Cl, in the presence of NaOH. In this method, the molar ratio of NaOH to diamide is in the range of 1.8 to 4.0. Upon completion of the Hoffman rearrangement reaction, the solution's pH is adjusted to about 9 to 14 via the addition of an amount of alkali base.

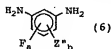


Applicants pointed out in their response dated August 5, 2008, that Masayoshi teaches certain compounds of formula (2), but does not teach any method of making them from diamides.¹ Indeed, the Examiner concedes that “Masayoshi does not teach a method for the production of a fluorinated aromatic diamine from corresponding diamide.” See the Office Action, page 4, lines 5-6. In other words, Masayoshi does not teach obtaining a diamine from its corresponding diamide, as required by claim 1, let alone, upon completion of the Hoffman rearrangement, adjusting the solution’s pH to about 9 to 14 via the addition of an amount of alkali base, as also required by claim 1.

Applicants now turn to Andrews. As correctly pointed out by the Examiner, this reference discloses preparing an isophthalic-based diamine from a corresponding diamide via the Hoffmann rearrangement. According to the Examiner, Andrews teaches that the NaOH/diamide ratio is 2.25, which is within the range of claim 1. See the Office Action, page 4, lines 11-13.

Applicants reiterate that Andrews discloses two methods which use the Hoffmann rearrangement for preparing an isophthalic-based diamine from its corresponding diamide. In the first method, the NaOH/diamide ratio was 2.25. This method resulted in a very low diamine yield of only **19%**. See page 416, lines 23-38. In the second method, characterized as “a more efficient preparation” in Andrews (see page 416, line 38), a NaOH/diamide ratio of about 1.56 was used, which resulted in a significantly increased diamine yield of **74%**. See page 416, lines 39-47. Applicants also reiterate that not only does Andrew’s “more efficient preparation” result in a higher yield, but also in a product with greater purity. Indeed, Andrews teaches that in the Hoffman rearrangement involving an isophthalic-based diamine appreciable amounts of by-products were produced by the heating of the bulk solution (i.e., the less efficient preparation of the first

¹ As correctly pointed out by the Examiner, Mayoshi teaches “a fluorinated phenylenediamine of ... formula (6) where Z” denotes a chlorine, bromine or iodine ... *a* denotes fluorine atoms bonded to a benzene ring, representing an integer of 0-4 ... and *b* denotes the number of “Z” bonded to a benzene ring, representing 0-4 ... the total of *a* and *b* ought to be invariably 4.” See the Office Action, page 2, line 17 through page 3, line 7; *a* and *b* have been italicized.



method), but the yield and purity were improved by using apparatus which ensured rapid heating and consumption of intermediates (i.e., the more efficient preparation of the second method). See page 414, lines 17-34. In sum, based on the teachings in Andrews, when a compound having both higher yield and purity is needed, a skilled artisan would prefer Andrews' more efficient preparation using a NaOH/diamide ratio of about 1.56. Note that Applicants previously presented the above arguments in their responses dated August 5, 2008 and December 8, 2008. Applicants reiterate that Andrews teaches away from using a NaOH/diamide ratio in the range of 1.8 to 4.0 recited in amended claim 1.

Applicants would also like to bring to the Examiner's attention that nowhere does Andrews teach or suggest that that upon completion of the Hoffmann rearrangement, the pH of the solution is adjusted to about 9 to 14 via the addition of an amount of alkali base.²

Applicants now turn to Hazen. As correctly pointed out by the Examiner, this reference "discloses a method for the production of a fluorinated aromatic diamine." See the Office Action, page 5, lines 11-19. As also correctly pointed out by the Examiner, "[the] Hazen process [the] molar ratio of [] NaOH/diamide [] is higher than 6." See page 6, lines 11-12. Indeed, nowhere in Hazen does it mention using a NaOH/diamide ratio in the range of 1.8 to 4.0, let alone, upon completion of the Hoffman rearrangement reaction, adjusting the solution's pH to about 9 to 14 via the addition of an amount of alkali base, both of which are required by amended claim 1. In fact, Hazen teaches that, upon completion of the reaction, the solution is treated with acetic acid so that its pH is adjusted to 7.5. See column 4, lines 64-65. In other words, Hazen actually teaches away from using an alkali base to adjust the pH of the reaction solution to about 9-14.

² As discussed above, in Andrews' first method, an excess of NaOH, i.e., 75 mmols to 12 mmols of diamide, is added at the beginning of the reaction. Comparative Example 1 of the Specification, demonstrates that when an excess amount of NaOH, i.e., 101.84 mmols of NaOH to 8.47 mmols of diamide, was added at the beginning of the reaction, the yield was very low, i.e., 16.4 %. This low yield can be attributed to the conversion of the fluoro groups on the diamide to hydroxyl groups and to formation of unwanted by-products. The Specification also teaches that if the NaOH to diamine ratio is below 1.8, the shortage of NaOH results in a lowering of the reaction yield. In sum, in the method of amended claim 1, it is important (i) not to have an excess of NaOH at the beginning of the reaction, (ii) not to have an NaOH/diamide ratio below 1.8, and (iii) to adjust pH of the solution with an alkali base only upon completion of the Hoffmann rearrangement.

Turning to Pfirmann, the Examiner asserts that this reference “teaches a method for the production of phenylenediamine which comprises steps of reacting fluorinated amide [with] NaOX [and] NaOH at a molar ratio of [] NaOH to [] diamide [] in the range of 1-30, **preferably 5-10.**” See the Office Action, page 4, lines 7-12; emphasis added. Applicants would like to point out that Pfirmann does not teach a method for the production of diamines. Rather, it teaches a method for the production of monoamines. See column 1, lines 11-16, and Examples 1-8. In particular, the Examiner refers to Example 2 of Pfirmann.

In Example 2, Pfirmann teaches that **1.5 mols of NaOH** and **0.164 mols of** 2, 6-difluorobenzamide, a monoamide, were used in the Hoffmann rearrangement. Pfirmann clearly teaches using an excess amount of NaOH. Indeed, the NaOH/monoamide ratio can be calculated as follows:

$$\begin{aligned} &(\text{mols of NaOH/mols of monoamide})-(\text{mols of NaOX used}) = \\ &(1.5 \text{ mols}/0.164 \text{ mols})-(1.23)=\mathbf{7.92}. \end{aligned}$$

For the determination of the numerical value 1.23, i.e., the mols of NaOX used, see footnote 3 below.³

In this example, Pfirmann teaches using a monoamide. Thus, to determine the NaOH/diamide ratio one must double the NaOH/monoamide ratio, i.e. $2 \times 7.92 = \mathbf{15.84}$. Given this teaching, it is clear that Pfirmann does not suggest using a NaOH/diamide ratio of 1.8-4.0, required by amended claim 1.

Admittedly, Pfirmann also teaches the broad NaOH range of 1-30. However, Applicants would like to bring to the Examiner’s attention that to determine whether there is motivation to select a species, i.e., the NaOH/diamide range of 1.8-4.0, from a

³ In this Example, 5 L of chlorine were used, i.e., 5 L/h x 1 hour. To calculate the NaOX/diamide ratio, if one assumes that chlorine is an ideal gas, the equation $PV=nRT$ can be used to calculate the number of mols of chlorine in 5L. This formula can be rearranged as follows: $V/n = RT/P$. In this formula, V is the volume of the ideal gas, n is the number of mols of chlorine found in the volume V, P is pressure, which is assumed to be 10^5 Pa (standard pressure), T corresponds to temperature, which is assumed to be 298K (or ambient temperature), and R is the ideal gas constant of 8.3145 L-KPa/mol-K. When all of the numbers are put into this formula, one can determine that 1 mole of chlorine, as an ideal gas, would occupy 24.8 L. Using this number, 5L of chlorine would have $5L/(24.8 \text{ mol/L})$ or 0.2016 mols, of chlorine. Thus, Pfirmann teaches using a NaOX/monoamine ratio of $0.2016/0.164$, i.e., 1.23.

prior art genus, i.e., the NaOH/monoamide range of 1-30, the Examiner must consider factors including the express teachings in the prior art. See MPEP 2144.08.II.4, which states in relevant part:

[t]o address this key issue [i.e., whether it would have been obvious to select the claimed species from the disclosed prior art genus], Office personnel should consider all relevant prior art teachings, focusing on [, among others, c]onsider[ing] the express teachings ... (emphasis added).

As correctly pointed out by the Examiner, Pfirmann expressly teaches that the preferred NaOH range is 5-10. Given that this range is for monoamides, the preferred NaOH/diamide ratio taught in Pfirmann is actually **10-20**. Nowhere in this reference does it suggest using an NaOH ratio of 1.8-4.0, as required by amended claim 1. Indeed, in view of the express teaching, or, more precisely, express teaching-away, a skilled artisan would have selected NaOH/diamide ratios in the range of 10-20 from the genus of ranges taught by Pfirmann, not 1.8-4.0. In turn, absent a selection of this range as the species, a skilled artisan would not have modified the NaOH/monoamine ratios of the Pfirmann method, thereby arriving at the NaOH/diamide ratios recited in claim 1.

In addition, Applicants point out that nowhere does Pfirmann teach or suggest that that upon completion of the Hoffmann rearrangement, the pH of the solution is adjusted to about 9 to 14 via the addition of an amount of alkali base. See footnote 2 *Supra*.

Finally, Applicants turn to the evidentiary reference Zengel. The Examiner relies on this reference to show that, though Pfirmann does not teach the use of a NaOH/diamide ratio of 6.0 in the Hoffmann rearrangement, Zengel teaches that the “optimal NaOH/diamide ratio [for this rearrangement] is 6:1.” See the Office Action, page 6, lines 13-15. It is clear that Zengel, like Pfirmann, does not suggest an NaOH/diamide ratio of 1.8-4.0.⁴ Note that this reference also does not teach or suggest

⁴ Applicants amendment to claim 1 have negated the Examiner's reliance on Zengel.

that, upon completion of the Hoffmann rearrangement, adjusting the pH of the solution to about 9 to 14 via the addition of an amount of alkali base.⁵

To conclude, none of Masayoshi, Andrews, Hazen, and Pfirmann, either alone or in combination, teaches or suggests the following two features of amended claim 1: (1) an NaOH/diamide ratio of 1.8-4.0 and (2) upon completion of the Hoffmann rearrangement, adjusting the pH of the solution to about 9 to 14 via the addition of an amount of alkali base.

For the reasons and facts set forth above, claim 1 is not rendered obvious by the cited references. Nor are claims 2-4 and 7-10, all of which depend from claim 1.

Applicants now turn to the rejection of independent claims 5, 6, 11, and 12. Claims 5 and 11 each cover a method for obtaining a polyamic acid while claims 6 and 12 each cover a method for obtaining a polyimide. These methods all require a NaOH/diamide ratio identical to that of claim 1, which is discussed above. Thus, for at least the same reasons and facts set forth above, claims 5, 6, 11, and 12 are also not rendered obvious by Masayoshi, Andrews, Hazen, and Pfirmann. Nor are claims 14 and 15, which depend from claims 5 and 6, respectively.

New Claims

Claims 16-20 has been added to cover particular embodiments of this invention. These new claims depend from claims 1, 5, 6, 11, and 12. For the same reasons set forth above, they are patentable over the cited references, either alone or in combination.

⁵ The Zengel method also differs from the method of claim 1 in that:

(1) it requires two stages (i): diamide reacts with chlorine under acidic conditions to produce dichloro-diamide (chlorination reaction) and (ii) the dichloro-diamide reacts with alkali or alkaline earth metal hydroxide to produce diamine, while the method of claim 1 takes place in a single stage, and

(2) the diamide used in the Zengel method does not have a halogen substituted benzene ring, while the diamide used in the method of claim 1 requires at least on halogenated group.

Applicant(s) : Shinji Nishimae
Serial No. : 10/561,048
Filed : December 14, 2005
Page : 17 of 17

Attorney Docket No.: 60004-109US1
Client Ref. No.: F 04-038-PCT/US/NS

CONCLUSION

It is believed that all of the pending claims have been addressed. However, the absence of a reply to a specific rejection, issue or comment does not signify agreement with or concession of that rejection, issue or comment.

In addition, because the arguments made above may not be exhaustive, there may be reasons for patentability of any or all pending claims (or other claims) that have not been expressed.


Finally, nothing in this paper should be construed as an intent to concede any issue with regard to any claim, except as specifically stated in this paper, and the amendment of any claim does not necessarily signify concession of unpatentability of the claim prior to its amendment.

The Petition for Extension of Time fee in the amount of \$490 is being paid concurrently herewith on the Electronic Filing System (EFS) by way of Deposit Account authorization.

Please apply any other charges to Deposit Account No. 50-4189, referencing Attorney Docket No. 60004-109US1.

Respectfully submitted,

Date: 6-25-09


Y. Rocky Tsao, Ph.D., J.D.
Attorney for Applicants
Reg. No. 34,053

Occhiuti Rohlicek & Tsao LLP
10 Fawcett Street
Cambridge, MA 02138
Telephone: (617) 500-2509
Facsimile: (617) 500-2499